Controlling the exchange interaction using the spin-flip transition of antiferromagnetic spins in $Ni_{81}Fe_{19}$ / α-Fe_2O_3

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Abstract

We report studies of exchange bias and coercivity in ferromagnetic $Ni_{81}Fe_{19}$ layers coupled to antiferromagnetic (AF) (0001), (11$\bar{2}$0), and (11$\bar{0}$2) α-Fe_2O_3 layers. We show that AF spin configurations which permit spin-flop coupling give rise to a strong uniaxial anisotropy and hence a large coercivity, and that by annealing in magnetic fields parallel to specific directions in the AF we can control either coercivity or exchange bias. In particular, we show for the first time that a reversible temperature-induced spin reorientation in the AF can be used to control the exchange interaction.

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The interaction between a ferromagnet (FM) and an antiferromagnet (AF) across an interface gives rise to an exchange bias (H_{ex}), i.e., a shift in the hysteresis loop as well as an enhanced coercivity (H_C) compared with a free FM material [1]. This exchange bias is fundamental to the operation of spin valve devices such as magnetic read-heads, non-volatile memories, and various sensors [2–4]. Despite considerable work by many groups over the past two decades, the origin of the exchange bias and the enhanced coercivity are still unclear [5–8]. One of the primary aims of the various models is the reconciliation of the experimentally observed values of H_{ex} and H_C with theoretical predictions. Mauri *et al*. [9] predict experimentally realistic values for H_{ex} on the assumption that a domain wall parallel to the surface is formed in the AF layer. The random field model of Malozemoff [10], by considering interface roughness, qualitatively explained exchange bias in compensated, but disordered, AF surfaces. Koon [11] demonstrated that it is possible for the FM layer to minimize its energy when it aligns perpendicularly to the AF easy axis; this type of perpendicular exchange coupling has become known as spin-flop coupling because of its similarity to the spin flop state of an AF material in a magnetic field. However, Schulthess and Butler [12] revealed that the spin-flop coupling alone cannot induce a unidirectional anisotropy, but instead gives rise to a uniaxial anisotropy which causes an enhanced coercivity. Experimentally, such spin-flop coupling has been observed in epitaxial FM/AF systems, such as $Ni_{80}Fe_{20}/Fe_{50}Mn_{50}$, Co/NiO, Fe_3O_4/CoO, and Fe/FeF_2 [13–18]. Nevertheless, a satisfactory understanding is not yet available because of complications at interfaces which include roughness, spin structure, and defects. Of particular relevance to the work presented here, Fitzsimmons *et al*. [18] have shown that exchange bias is dependent on the in-plane crystalline quality, and hence the net spin configuration at the interface, of an AF layer. The aim of the experiments reported here was to investigate the exchange bias and coercivity in a system in which the interfacial AF spin configuration could be controlled and changed without modifying the structural properties of the AF/FM interface.

Hematite α-Fe_2O_3 is potentially attractive for exchange biased applications because of its high Neel temperature ($\sim$680 ^{0}C) [19,20]. Bulk α-Fe_2O_3 undergoes an unusual temperature-

controlled transition between two AF spin configurations - the so-called spin-flip (Morin) transition at T_M ∼260 K. Thus the spin configuration of a single-crystal α-Fe_2O_3 surface not only depends on the direction of that surface, but can be changed by altering the temperature. It has already been identified as an ideal system in which to study exchange bias in general and spin flop in particular [21].

It has been shown that epitaxial α-Fe_2O_3 films on α-Al_2O_3 substrates have spin-flip transition temperatures which depend on the crystal orientation [22,23]: the T_M of $(11\bar{2}0)$ α-Fe_2O_3 is similar to that of the bulk material, while (0001) α-Fe_2O_3 films do not show a spin-flip transition above 2.5 K. In contrast, the spin-flip transition of $(1\bar{1}02)$ α-Fe_2O_3 films is increased to about 400 K; in this case the AF spins lying within the film plane above T_M flip to the out-of-plane direction below T_M. These changes are associated with lattice strain caused by the lattice mismatch between α-Fe_2O_3 and α-Al_2O_3 (∼5.5 %).

In this Letter, we report exchange bias and coercivity in FM layers coupled with epitaxial α-Fe_2O_3 layers. In contrast to previous experiments which have compared the exchange interaction associated with different *fixed* spin orientations associated with different AF crystal faces [16], we show for the first time that a change of AF spin orientation across a *single* interface is directly reflected in a modified exchange interaction.

Epitaxial α-Fe_2O_3 films were grown on (0001), $(11\bar{2}0)$, and $(1\bar{1}02)$ α-Al_2O_3 substrates by pulsed laser deposition (PLD) with a substrate temperature of 700 ^{0}C and oxygen pressure of 20 mTorr. In order to fabricate the films under identical conditions, three substrates were loaded side by side for simultaneous deposition. The 50 nm thick α-Fe_2O_3 films were transferred into an ultra high vacuum dc sputtering chamber and a 5 nm $Ni_{81}Fe_{19}$(NiFe) film was deposited in a magnetic field of 250 Oe at 295 K.

X-ray diffraction (XRD) measurements showed that all three α-Fe_2O_3 growth directions yielded films with excellent crystallinity: a full width half maximum of $< 0.08^0$ and $\sim 0.9^0$ in the rocking curve and in the ϕ scan, respectively. The *rms* surface roughness measured by atomic force microscopy was about 0.5 nm in each case. Therefore, effects caused by extrinsic factors such as roughness and defects should be virtually same for all three samples, and

thus differences in the H_{ex} and the H_C should depend only on the spin structure of α-Fe_2O_3 at the surface. The strains observed in our films are similar to those reported by Fujii et al. [22] and so we expect a similar change in the Morin temperature.

The NiFe/α-Fe_2O_3 samples were measured in a variable temperature vibrating sample magnetometer (VSM). Fig. 1 shows the temperature-dependent magnetic hysteresis loops of as-prepared samples for the different crystal orientations; each panel shows magnetization data collected for two orthogonal in-plane directions (see Fig. 3). For the (0001) orientation, the hysteresis loop is essentially independent of the temperature and the in-plane field direction: it shows minimal H_{ex}, and a coercive field of about 22 Oe. In contrast, the $(11\bar{2}0)$ α-Fe_2O_3 system shows large changes between 25 and 295 K. Fig. 2 shows how the magnetization and saturation field of $(11\bar{2}0)$ α-Fe_2O_3 / NiFe for two in-plane directions depends on temperature; it is clear that the easy axis rotates by 90^0 over the temperature range of the experiment. Finally, hysteresis loops of the NiFe on $(1\bar{1}02)$ α-Fe_2O_3 / NiFe for two in-plane directions at 295 K are virtually identical except a slight shift, but at 380 K there is a clear difference for the two in-plane directions, i.e. easy and hard magnetization axes.

Fig. 3 shows the crystal-direction dependence of the surface spin configuration of α-Fe_2O_3 [22–24]; the NiFe spin will always lie within the film plane because of large shape anisotropy. For spin-flop coupling to give rise to a strong uniaxial anisotropy, the FM spins must align perpendicular to the AF spins [12]. Accordingly, if the spin-flip transition of α-Fe_2O_3 at T_M results in a change of the in-plane spin direction, this should be reflected by a change in the easy axis of the NiFe. This expectation is consistent with our results for the $(11\bar{2}0)$ and $(1\bar{1}02)$ α-Fe_2O_3, as seen in Figs. 1 and 2: the 90^0 rotation of the easy axis of NiFe on the $(11\bar{2}0)$ α-Fe_2O_3 is associated with the spin-flip transition of α-Fe_2O_3 from the ab plane to the c-axis (albeit associated with a reduced T_m which is typical for 40-50 nm length scales in α-Fe_2O_3 [22,25]), while the FM spins on $(1\bar{1}02)$ α-Fe_2O_3 at room temperature have no preferential orientation because all in-plane directions equally satisfy a spin-flop coupling condition. Upon warming, however, a preferential direction appears within the plane as the AF spins flip to one of the in-plane directions. [22] The substantially lower coercivity for the

NiFe on (0001) α-Fe_2O_3 is also consistent with this picture since it has an uncompensated surface at all temperatures, and so cannot generate spin-flop coupling. It is important to note that this uncompensated surface gave negligible exchange bias following any annealing procedure in contradiction to simple models for such systems; this may be a consequence of the small, but finite roughness in any practical sample. Although this may appear surprising, it is consistent with previous results in the exchange biased epitaxial system Fe/FeF_2, which showed zero exchange bias for an uncompensated AF surface [16].

If the intrinsic anisotropy energy of a FM layer is negligible, the total energy per unit area in an exchange coupled FM/AF system can be expressed as [26]

$$E = -J_1 cos\theta - J_2 sin^2\theta + K_{AFM} sin^2\phi \tag{1}$$

where J_1 and J_2 are respectively a direct (parallel) coupling constant and a spin-flop (perpendicular) coupling constant; θ and ϕ are the angles between the FM spin and the AF spin directions, and the AF spin and the AF anisotropy axis; K_{AFM} is the anisotropy constant of the AF layer. The lowest energy state is thought to be a spin flop like state ($\theta = 90^0$, $\phi = 0^0$ (see $T > T_m$ in Fig. 3(b),(c)). The form of the spin flop coupling is comparable with the classical uniaxial anisotropy energy, and thus the coercivity is mainly dependent on the second term of (1). If we associate exchange bias with a domain wall formed in the AF layer [7,10,27], its stability is determined by a competition between a decrement of direct coupling energy and an increment of the AF anisotropy energy. From equation (1), we expect that a magnetic field annealing (MFA) process perpendicular to the AF spin direction will stabilize the spin-flop coupling, and in turn it will enhance the coercivity. On the other hand, a MFA process parallel to the AF spin direction should induce an exchange bias because it enhances the direct coupling and suppresses the spin flop coupling.

We performed a series of experiments in which MFA was performed under 10 kOe for 15 minutes at 200 ^{0}C, and the samples were cooled down to room temperature in the magnetic field. The $(11\bar{2}0)$ and $(1\bar{1}02)$ α-Fe_2O_3 films with a compensated surfaces showed distinctive MFA effects; Fig. 4 shows the hysteresis loops of NiFe on (1120) α-Fe_2O_3 before and after

MFA. When the MFA was performed perpendicular to the AF spin direction (Fig. 4(d)), the exchange bias showed no change, but the coercivity approximately doubled (Fig. 4 (b)). In contrast, H_{ex} of order 80 Oe was induced along the hard axis when the MFA was performed parallel to the AF spin direction (Fig. 4 (c)). A similar exchange bias along the hard axis has been observed in the epitaxial Fe/$FeFe_2$ system.[17,18] Thus MFA with a configuration of Fig. 4(e) enhances the direct coupling in the equation (1), and in turn it induces the H_{ex} along the hard axis.

Finally, we applied MFA to the NiFe on $(1\bar{1}02)$ α-Fe_2O_3. A large exchange bias of 80-100 Oe was induced by the MFA along all directions within the plane, as seen in Fig. 5. If $(1\bar{1}02)$ α-Fe_2O_3 has the ideal spin structure of Fig. 3(c) below T_M, the NiFe should have shown no exchange bias because the spin flop coupling is dominant. On the contrary, the large exchange bias suggests that the AF spin-flip transition to the out-of-plane direction during the cooling process of MFA is frustrated at the interface because of in-plane FM spins. In Figure 5 (c), the temperature dependent exchange bias clearly shows an anomaly at T_*, which agrees with the T_M of $(1\bar{1}02)$ α-Fe_2O_3 in previous report [22]. We conjecture that the MFA assisted by FM spins enhances the direct coupling leading the formation of a domain wall in the AF layer, and thus the exchange bias is induced along all in-plane directions.

The magnitude of exchange bias depends on the time and the temperature of the MFA; the high Neel temperature of α-Fe_2O_3 might be expected to limit the effectiveness of low-temperature MFA. In the present study, the largest value of H_{ex} at room temperature was about 100 Oe for the NiFe/ $(1\bar{1}02)$ α-Fe_2O_3 film. If we simply assume that the effective interface exchange energy corresponds to a net direct exchange energy, the energy relation is $J_1 = M_F H_{ex} t_F$, where M_F and t_F are the saturation magnetization and thickness of the FM layer, respectively. Therefore, the estimated direct coupling constant J_1 is about 0.04 erg/cm^2 at room temperature; this value is similar to that in NiFe/NiO [28]. On the other hand, the maximum difference of the coercivity between NiFe/ (0001) α-Fe_2O_3 film and NiFe/ $(11\bar{2}0)$ or $(1\bar{1}02)$ α-Fe_2O_3 film was about 180 Oe at room temperature. Because such

a difference of the coercivity is mainly due to the spin-flop coupling, the spin-flop coupling J_2 can be estimated to be roughly 0.07 erg/cm^2.

In summary, we have shown for the first time that a change of spin orientation in an AF material is directly reflected in a modified exchange interaction. We have also demonstrated that magnetic field annealing parallel to specific directions in the AF can alternatively modify either the coercivity or the exchange bias, in agreement with the Schulthess and Butler [12] and Malozemoff models [10] respectively, demonstrating that these pictures of exchange bias are not mutually exclusive, but are part of a wider picture. This systematic study should improve the understanding of fundamental origins of exchange bias and the coercivity in exchange biased systems.

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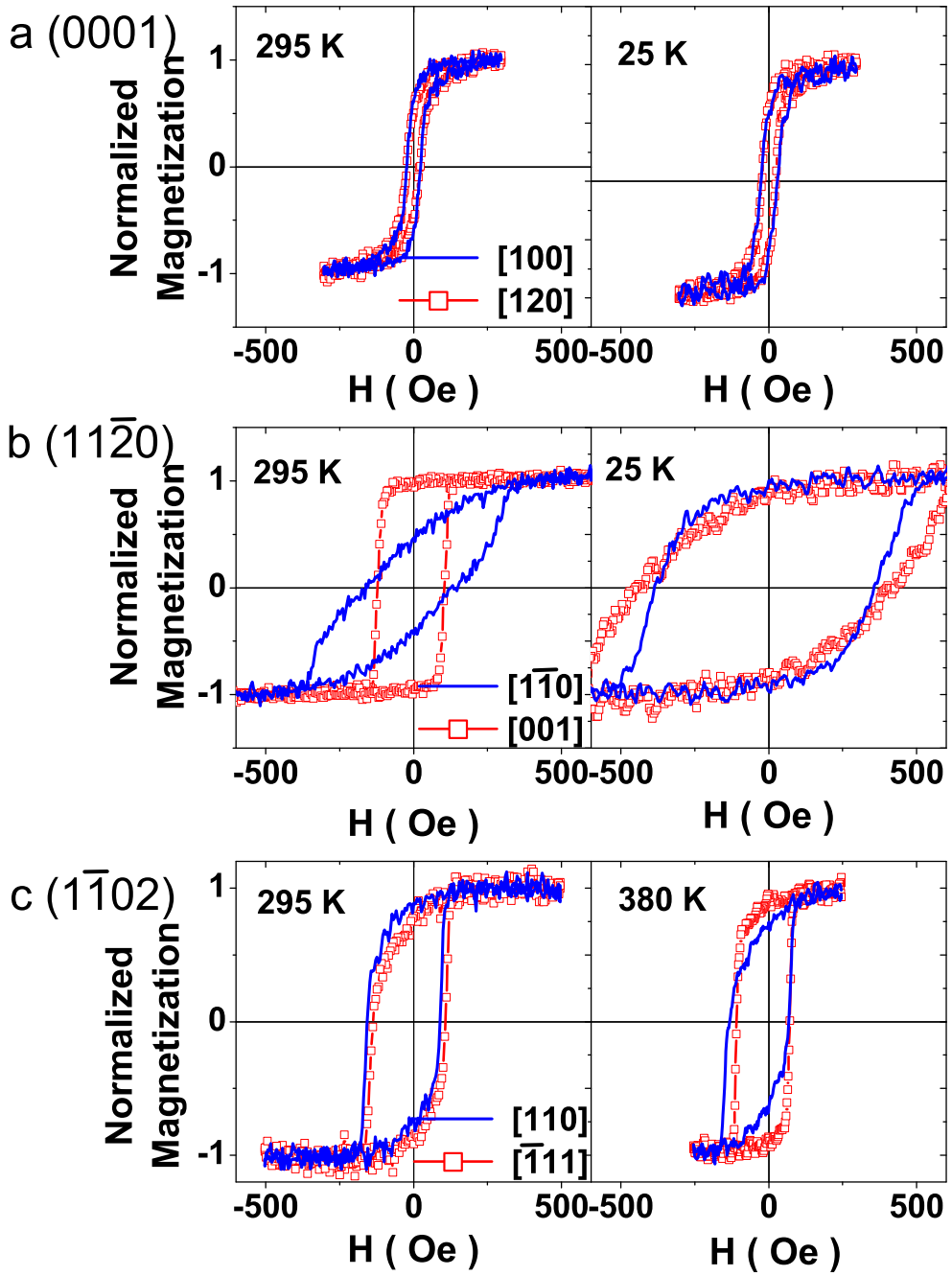

FIG. 1. The magnetic hysteresis loops of as-prepared $Ni_{81}Fe_{19}$ on (a) (0001) α-Fe_2O_3, (b) (11$\bar{2}$0) α-Fe_2O_3, and (c) (1$\bar{1}$02) α-Fe_2O_3 at several temperatures.

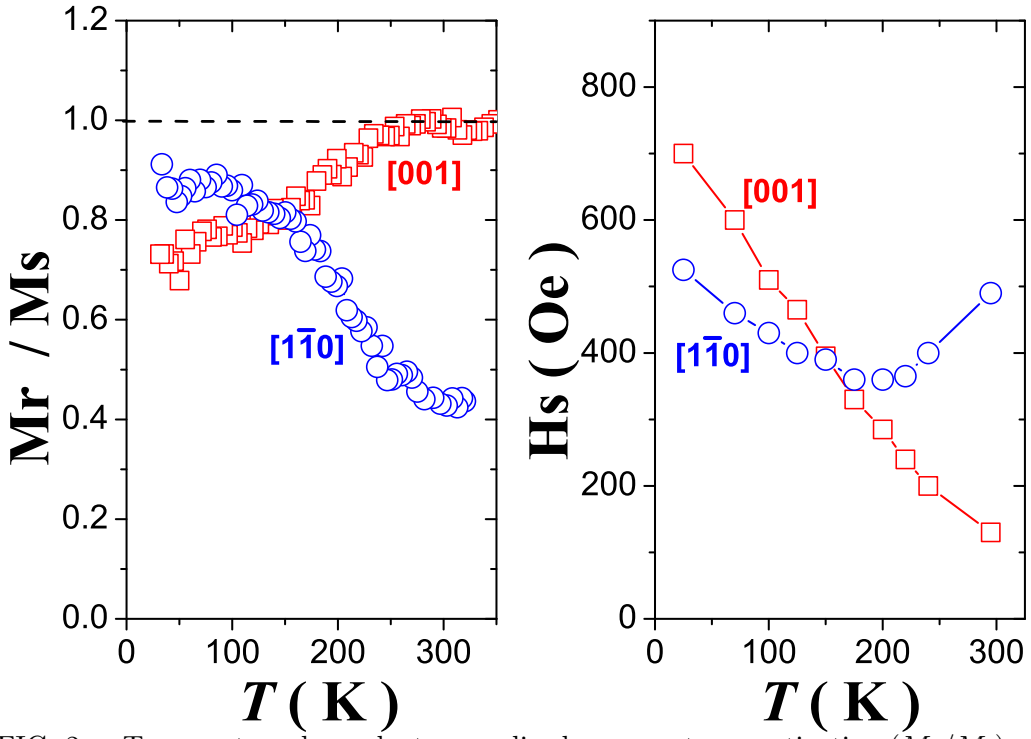

FIG. 2. . Temperature dependent normalized remanent magnetization (M_r/M_s) and saturation field (H_s) of as-prepared $Ni_{81}Fe_{19}$ on $(11\bar{2}0)$ α-Fe_2O_3 with the in-plane direction.

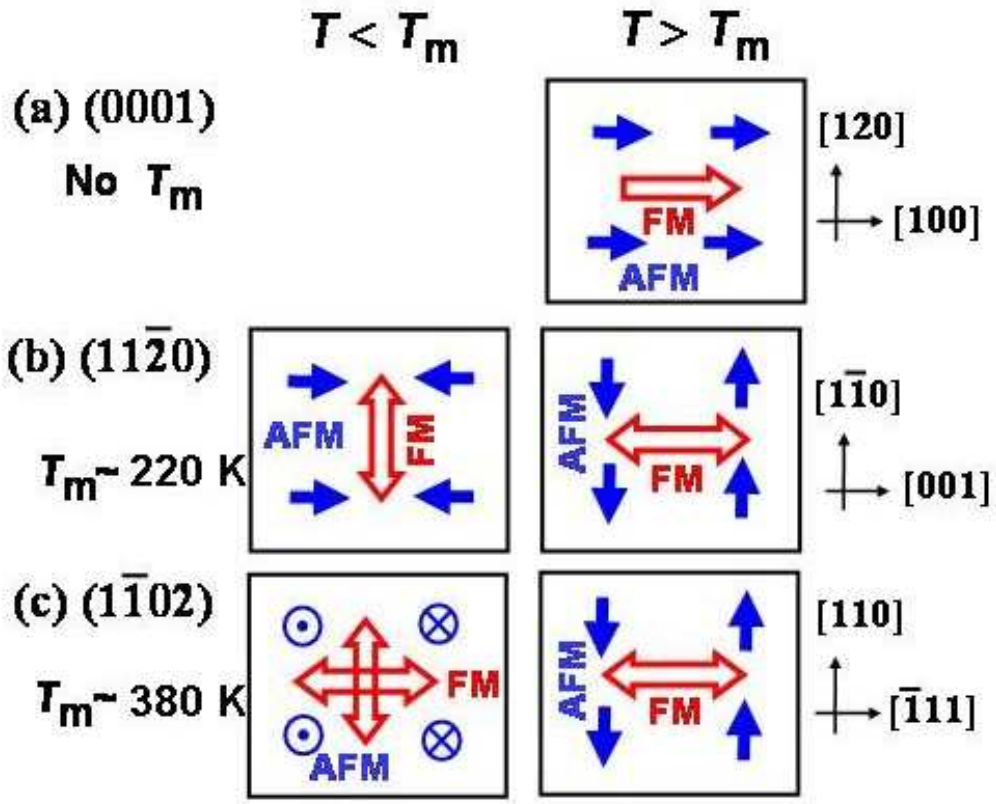

FIG. 3. Schematic surface spin structures of α-Fe_2O_3 (a) on (0001) α-Al_2O_3, (b) on (11$\bar{2}$0) α-Al_2O_3, and (c) on (1$\bar{1}$02) α-Al_2O_3.

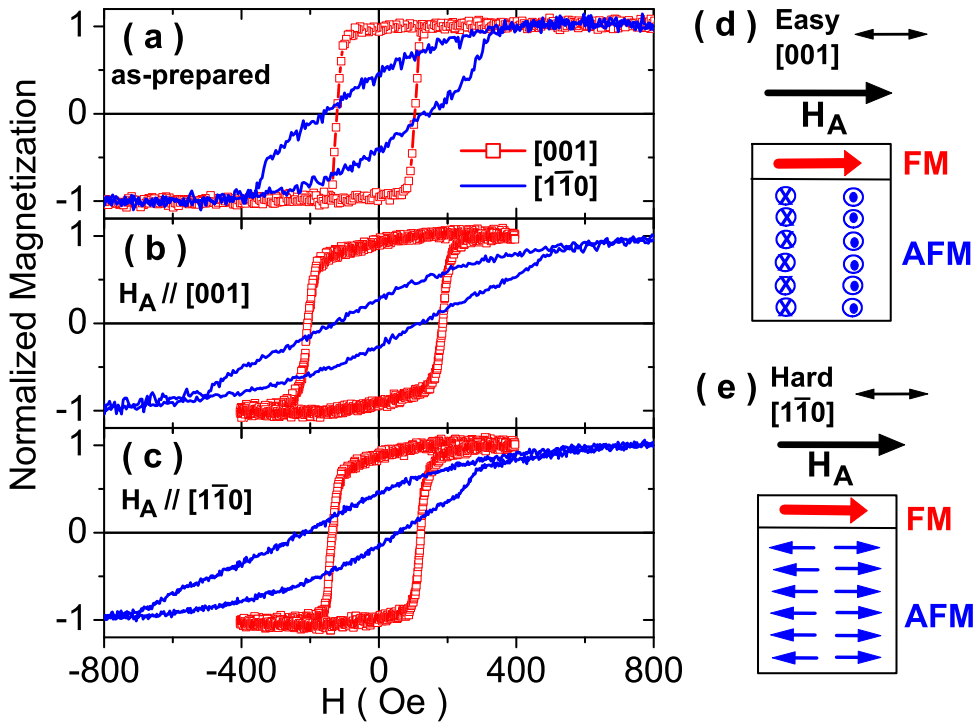

FIG. 4. The room temperature magnetic hysteresis loops of $Ni_{81}Fe_{19}$ on $(11\bar{2}0)$ α-Fe_2O_3 (a) without MFA, (b) with MFA perpendicular to the AF spin direction, and (c) with MFA parallel to the AF spin direction. Two schematic MFA configurations were also displayed in (d) and (e). Here, H_A means the annealing magnetic field.

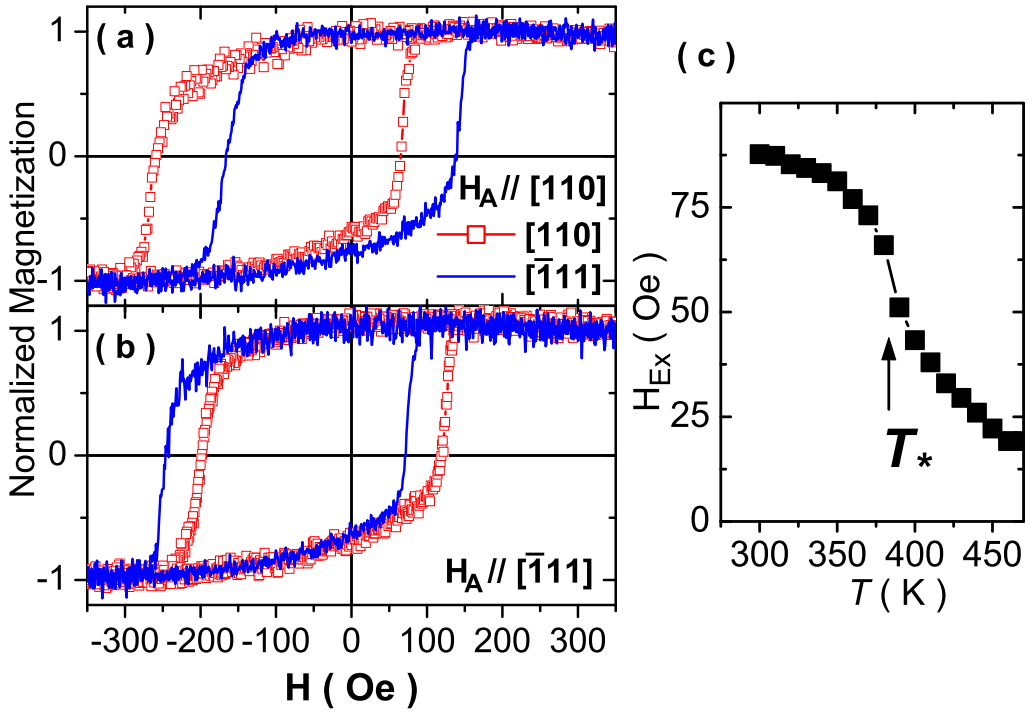

FIG. 5. The room temperature magnetic hysteresis loops of $Ni_{81}Fe_{19}$ on $(1\bar{1}02)$ α-Fe_2O_3 (a) with MFA along the [110] direction, and (b) with MFA along the $[\bar{1}11]$ direction. The temperature dependent exchange bias H_{Ex} was displayed in (c).